Ecotoxicology and Environmental Safety 166 (2018) 251–258

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

Quantitative screening level assessment of human risk from PCBs released in glacial meltwater: Silvretta Glacier, Swiss Alps

K.R. Miner[a,f], C. Bogdal[b], P. Pavlova[c,d], C. Steinlin[b,e], K.J. Kreutz[a]

[a] Climate Change Institute, University of Maine, Orono, ME 04469, USA
[b] Institute for Chemical and Bioengineering, ETH Zurich, CH-8093 Zurich, Switzerland
[c] Agroscope, Schloss 1, CH-8820 Wädenswil, Switzerland
[d] Analytical Chemistry Group, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
[e] EBP Schweiz AG, CH-8702 Zollikon, Switzerland
[f] ERDC-Geospatial Research Laboratory, Alexandria, VA 22315, USA

ARTICLE INFO

Keywords:
Risk assessment
Glacier
Water
Persistent pollutants
Climate change
Toxicology

ABSTRACT

Persistent organic pollutants (POPs) are entrained within glaciers globally, reemerging in many alpine ecosystems. Despite available data on POP flux from glaciers, a study of human health risk caused by POPs released in glacial meltwater has never been attempted. Glaciers in the European Alps house the largest known quantity of POPs in the Northern Hemisphere, presenting an opportunity for identification of potential risk in an end-member scenario case study. With methodology developed by the US Environmental Protection Agency (EPA), we provide a regional screening level human risk analysis of one class of POPs, polychlorinated-biphenyls (PCB) that have been measured in melt waters from the Silvretta Glacier in the Swiss Alps. Our model suggests the potential for both cancer and non-cancer impacts in residents with lifetime exposure to current levels of PCB in glacial meltwater and average consumption of local fish. For residents with an abbreviated 30-year exposure timeframe, the risk for cancer and non-cancer impacts is low. Populations that consume higher quantities of local fish are predicted to be at a greater risk, with risk to lifetime consumers higher by an order of magnitude. Based on the results of our screening study, we suggest that local government move to the next step within the risk assessment framework: local monitoring and management. Within the Alps, other glacial watersheds of a similar size and latitude may see comparable risk and our model framework can be adapted for further implementation therein.

1. Introduction

Despite being banned by the multinational Stockholm Convention agreement in 2004, long-range atmospheric transport has made some persistent organic pollutants (POPs) ubiquitous in the environment (Bogdal et al., 2009; Simonich and Hites, 1995; Villa et al., 2001).

In Arctic and high alpine environments, POP sorption to snow crystals leads to deposition and entrainment in glaciers, resulting in the long term storage of legacy chemicals in glacial ice (Blais et al., 2001; Miner et al., 2018, 2017; Pavlova et al., 2016). POPs also accumulate at enhanced rates in high-altitude, high-snowfall alpine environments proximal to areas of use (Daly and Wania, 2004; Dong et al., 2017, 2015; Pavlova et al., 2014). Glacial entrainment of POPs has been documented in mountain ranges within China, Svalbard, Canada, Switzerland and Italy, with the highest concentrations found in the European Alps due to a latitudinal atmospheric gradient (Lafrenière et al., 2006; Miner et al., 2017; Wang et al., 2008). The central location and high altitude of the Swiss Alps concentrates pollution input from both local and distal sources, with glacial POP and PCB concentrations the highest of any sampled to date in Europe or the Arctic (Ferrario et al., 2017; Miner, 2018).

Though POPs have been identified in glaciers globally, long-term human risk from uptake of fish and meltwater from the glacial watershed is unknown. Even at low concentrations, POPs in the environment have the potential to create health problems in downstream populations with long term exposure (Iszatt et al., 2015; Jaacks and Staimez, 2015; Ngwa et al., 2015).

Since 1975, the U.S. Environmental Protection Agency (EPA) has developed protocols for screening human cancer and non-cancer risk from chemicals at varying scales (EPA, 2017, 2003, 2001a; EPA et al., 2009). Applying a regional, multi-variable linear tool that utilizes both a conceptual and quantitative framework is the most comprehensive way to survey the potential for human toxicological risk within a larger glacial watershed. To establish a risk baseline we use observed

E-mail address: kimberley.miner@maine.edu (K.R. Miner).

https://doi.org/10.1016/j.ecoenv.2018.09.066
Received 23 July 2018; Received in revised form 12 September 2018; Accepted 15 September 2018
Available online 28 September 2018
0147-6513/ Published by Elsevier Inc.

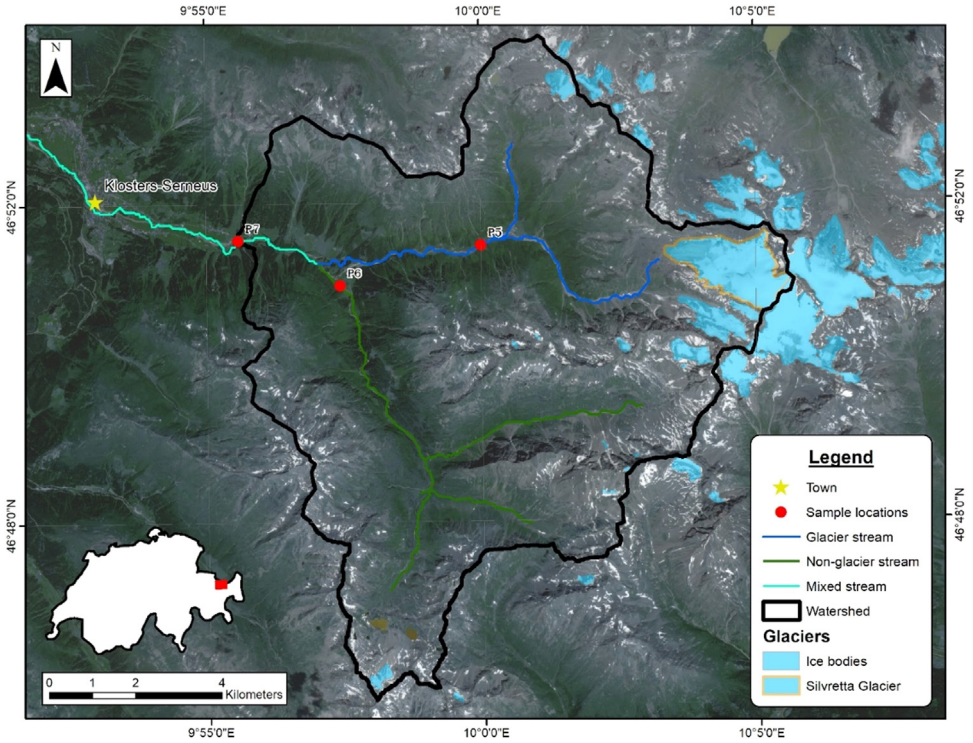

Fig. 1. Silvretta Glacier watershed with Silvretta Glacier outlined in orange. P5 is the sample location fed directly by glacial meltwater, P6 is the sample location for non-glacial samples and P7 is the sample location for the mixed stream. Klosters-Serneus is the closest town to the Silvretta Glacier and is indicated by a yellow star.

concentrations in a well-researched, endmember location, the Silvretta Glacier in the Swiss Alps.

The combination of ice cores, water samples, lake cores and glacial flow models from the Silvretta glacier (Fig. 1) makes it a glacier with one of the most complete data sets of glacial POP movement over time, providing a baseline for hazard identification (Pavlova et al., 2016, 2014). Therefore, the goal of this study is to provide the first assessment of population-level human risk from the uptake of PCB pollutants in glacial meltwater. While this study is site specific, the need to understand the potential risk to humans from pollutants in glacial meltwater is broadly applicable to numerous glaciated ranges.

2. Materials and methods

2.1. Silvretta data and exposure assessment

The field of glacial POP toxicology is in its infancy, characterized by a dearth of data. To account for this and to establish a risk baseline, we incorporate published research from the most-studied glacier in the Swiss Alps (Pavlova et al., 2016, 2014; Steinlin et al., 2016). Silvretta glacier, with an area of 3 km

^{2}, is a temperate alpine glacier located in the Central Eastern Range of the Swiss Alps where yearly ice melt begins in April and ends in December (Fig. 1) (Pellicciotti et al., 2010). Fourteen meltwater samples taken at three locations (P5,P6,P7-Fig. 1) in the glacial watershed during the summer months of two years were analyzed for 6 representative PCB congeners, and characterize the peak yearly glacial meltwater contribution to the watershed (Pavlova et al., 2016). While the bulk water sample data for the proglacial stream (P5: 73 pg/L-*Sept. 2013*; 52 pg/L-*Oct. 2013*) and non-glacial stream (P6: 51 pg/L-*Sept. 2013*; 55 pg/L-*Oct. 2013*) are similar, samples from the mixed stream where glacial and non-glacial water sources combine (P7: 100 pg/L-*Sept. 2013*; 100 pg/L-*Oct. 2013*) show an elevated PCB concentration indicative of the mixture of both sources (Pavlova et al., 2016; Steinlin et al., 2016). Though these concentrations are specific to the Silvretta watershed, results are within the observed range for PCB samples taken throughout the Alps within both glacially-fed streams and lake waterbodies over the past 10 years (Bogdal et al., 2010; Quadroni and Bettinetti, 2017; Schmid et al., 2011; Villa et al., 2001).

For this risk assessment, the upper limit PCB water concentration data was derived from the highest sample concentrations in the mixed stream bulk samples (P7–100 pg/L) with the average concentrations determined using the mean of all mixed stream data (P7–57 pg/L, averaged from 4 samples from 2012 and 2013). This allows us to identify the potential for risk under the highest observed concentration, as well as the median concentration that may more closely mirror the below glacier watershed. Though glacial melt peaks during summer months and dwindles during the winter, chemical concentrations in proglacial lakes and streams will continuously reflect the accumulation of multi-year chemical release and environmental partitioning (Grimalt et al., 2001; Mackay, 2001; Schmid et al., 2011). In accordance with standard practice, these concentrations were multiplied by a factor of 5 to account for the 30 representative PCB congeners frequently found in the environment (Glüge et al., 2017; Pavlova et al., 2016, 2014; Steinlin et al., 2015).

The below glacier watershed encompasses a variety of potential PCB exposure routes for humans, including the consumption of glacial water or resident fish, direct contact with the skin and inhalation of volatilized chemicals (Fig. 2a, b) (EPA, 1999; Bettinetti et al., 2008; Czub and McLachlan, 2004a; Czub and McLachlan, 2004b; Ren et al., 2016; Schmid et al., 2007). Compared to some industrial waterways such as the Hudson River, Silvretta Glacier meltwater exhibits relatively low concentrations of PCBs (EPA, 2001b). Accordingly, we identified that exposure through skin contact and inhalation posed extremely low risk during the initial exposure assessment and was thus eliminated from the quantitative screening model.

Within the Swiss Alps, high altitude, proglacial lakes immediately below the glacial terminus often contain fish, increasing the potential for bioaccumulation in fish tissue from waterborne chemicals (EPA, 2015; Mackay et al., 2001; Mackay and Patterson, 1990; Ren et al.,

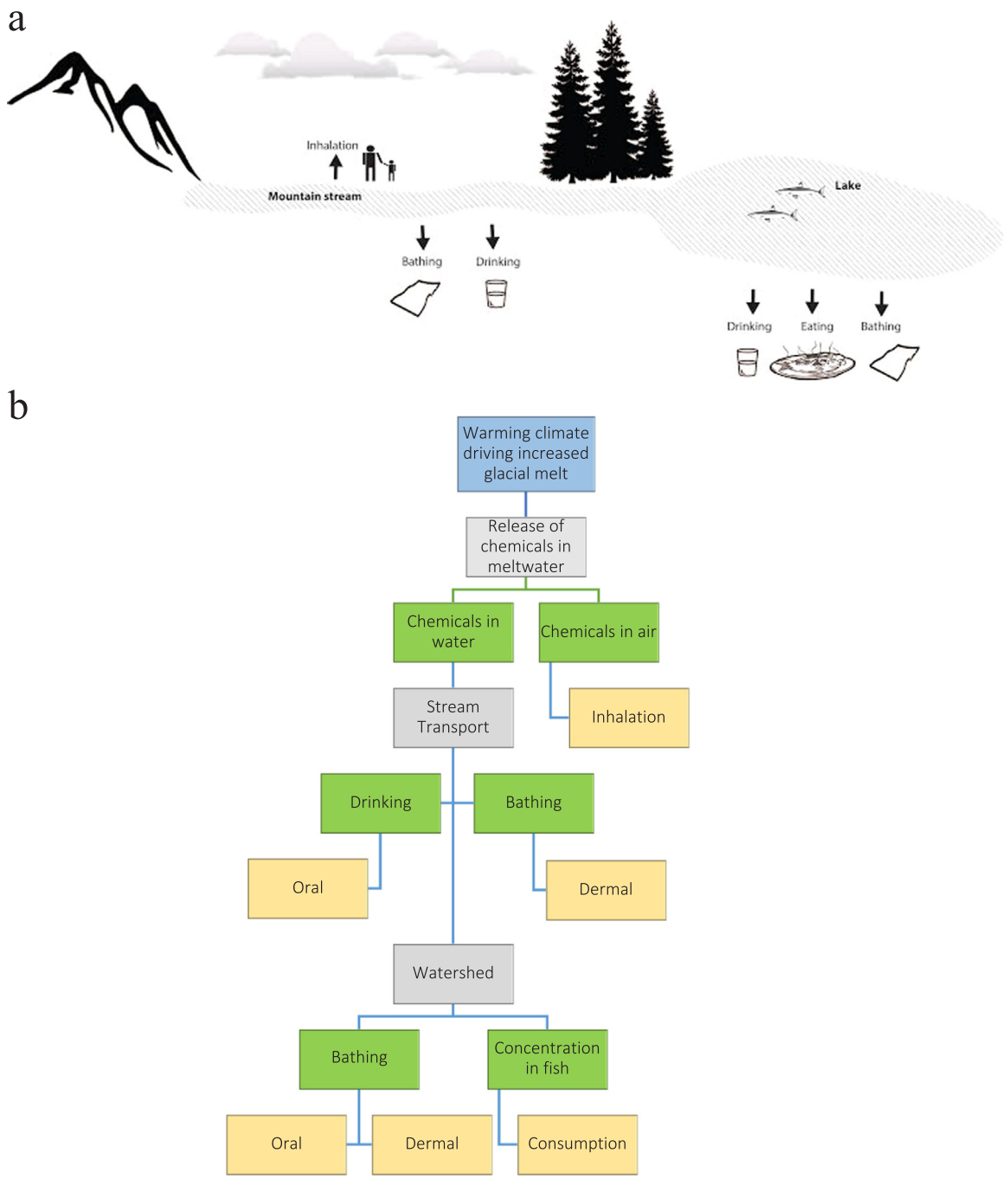

Fig. 2. a. Conceptual exposure assessment model of potential PCB human uptake routes from glacial meltwater in the Silvretta proglacial watershed. b. Conceptual exposure assessment flow model of potential PCB human uptake routes in the Silvretta proglacial watershed. Grey indicates ecosystem transport routes of PCB, green indicates human exposure sources and yellow indicates the type of human exposure.

2016; Schmid et al., 2007). However, the proglacial lake immediately below Silvretta Glacier is frozen through in the winter and no fish reside there. Therefore, to better represent a high-risk scenario integrating fish consumption, we utilized concentration data from the lower watershed mixed stream (P7) that contains endemic Salmonidae fish and is fed by both glacial and seasonal sources (Fig. 1). We subsequently incorporated bioaccumulation factors developed for PCB uptake in salmonid species to estimate storage in fish tissue (EPA, 2015; Office of Environmental Health Hazard Assessment, 2012). Such bioaccumulation in fish can increase PCB concentrations by up to 6 orders of magnitude relative to that of the surrounding water, making fish the greatest potential contributor of risk to human health in the local environment (EPA, 1999; Dewailly, 2006; Kallenborn, 2006; Quadroni and Bettinetti, 2017).

2.2. Quantitative screening model

The goal of applying a regional, multi-variable, linear screening level model is to determine if human health risk from consumption of glacial meltwater or fish could be high enough to necessitate additional analysis or mitigation on an individual or sub-group scale (EPA, 2003, 2001c, 1999).

Utilizing risk assessment framework guidelines from the EPA, we have incorporated average consumption data into published model framework that identifies both cancer and non-cancer risk (EPA, 2001a).

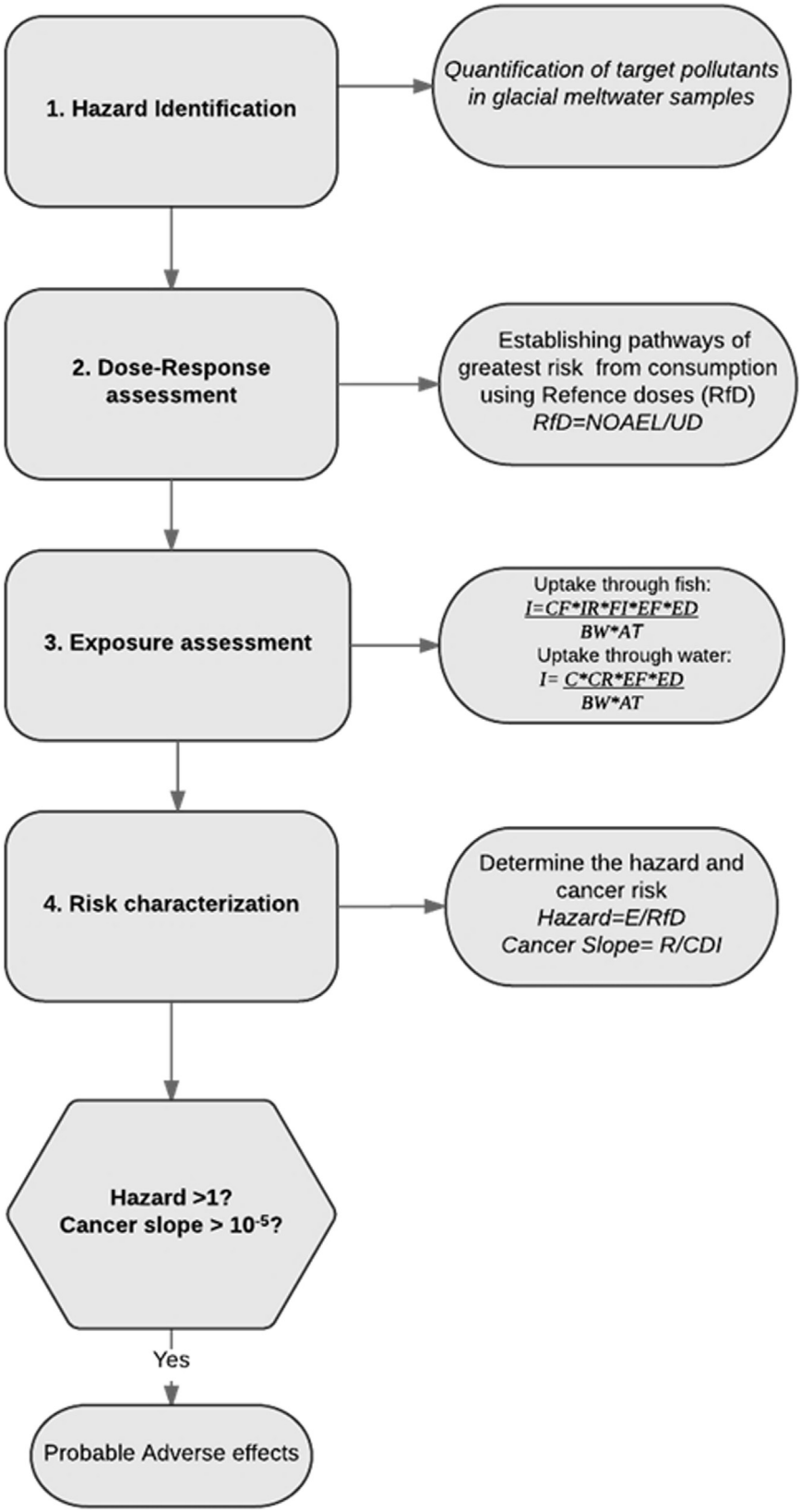

Fig. 3. Flow diagram of EPA quantitative screening risk assessment methodology utilized in this study as adapted from Zhai et al., 2017.

Initial steps are predicated on retrieving PCB concentration data, determining human uptake pathways (Fig. 2a,b), doses and duration (Fig. 3) ((EPA), 1999). Within this framework we incorporate the EPA Guidelines for Exposure Assessment and Framework for Cumulative Risk Assessment (Fig. 3) (EPA, 2015, 2003, 1999) in combination with fish consumption data from the Switzerland Federal Statistical Office (SFSO) (Statistical Office, 2017, 2016) and bioaccumulation factors from the California Office on Environmental and Health Hazard

Table 1
Cancer and non-cancer risk summary for 6% annual domestic fish consumption (population average- 0.0013 kg/day). Mixed stream (P7) data is for both the average (2.8×10^{-7} ng/L) and upper limit (5.0×10^{-7} ng/L) PCB concentration. Threshold for risk is 1 in non-cancer (NC) screening and 1×10^{-5} in cancer screening, with numbers over threshold indicated in bold.

Point Estimate Cancer Risk Summary, median fish consumption

Pathway	Cancer Median Risk	Cancer Highest Risk	NC Median Risk	NC Highest Risk
Fish Consumption				
Adult	**2.1 × 10–5**	**3.7 × 10–5**	0.23	0.41
Child	**7.3 × 10–5**	**1.3 × 10–4**	0.8	**1.4**
Water Consumption				
Adult	6.9 × 10–9	1.2 × 10–8	7.6 × 10 −5	1.3 × 10–4
Child	3.9 × 10–8	7.0 × 10–8	4.3 × 10–4	7.7 × 10–4
Both Pathways				
Adult	**2.1 × 10–5**	**3.7 × 10–5**	0.23	0.41
Child	**7.3 × 10–5**	**1.3 × 10–4**	0.8	**1.5**

Assessment (OEHHA) (Office of Environmental Health Hazard Assessment, 2012). These bioaccumulation factors calibrate chemical uptake in fish tissue by utilizing the mean percentage of muscle lipid tissue, octanol-water partition coefficient, chemical species and characterization of surrounding media (see Supplementary information for specific model variable inputs) (EPA, 2016; Mackay and Patterson, 1990; Office of Environmental Health Hazard Assessment, 2012). Fish species native and endemic to Switzerland include numerous members of the Salmonidae family, a fish consumed by local populations, for which we specifically calibrated our bioaccumulation factors (Froese, 2017). SFSO statistics indicate that the mean domestic fish consumption for Swiss residents is 6% (0.0013 kg/day) of total annual fish consumption (0.022 kg/day) (Statistical Office, 2017). Domestic fish for consumption are generally found in rivers and lakes with water inputs of a mixture of snow, rain and glacial meltwater, analogous to the mixed stream from which we derive concentration data. This allows us to use the Silvretta watershed as an initial case study for a model that could subsequently be applied to similar sized glaciers with resident fish populations throughout the region.

Human population residence time, water consumption and body weight are standardized in the EPA methodology based upon population averages (EPA, 2005, 1999; Phillips and Moya, 2013). The EPA Integrated Risk Information System (IRIS) is used to identify the cancer and non-cancer risk thresholds determined by laboratory-based toxicology analysis (EPA, 2017). For children, the model incorporates lifetime (70 year) consumption of glacial meltwater and domestic fish, whereas for adults, a 30-year timeframe is standard.

The risk and hazard assessment entails two main equations, one for cancer risk and one for non-cancer hazard. For cancer risk computations, oral intake of polluted meltwater or fish species utilize the same equation:

$$CDI = C*CR*EF*ED/BW*AT \quad (1)$$

Where CDI represents chronic daily intake, C is concentration of the pollutant, CR is the rate of contact through consumption, EF is how many days in a year this contact is made, and ED is the number of years contact is made. BW represents body weight which is also a proxy for age and follows the average population weights published by EPA (1999), AT is the averaging time which represents years in terms of days. The equation gives the result in terms of a dosage: mg/kg/day (CDI).

This number is then multiplied times the laboratory derived slope factor for cancer (SL) or upper bound limit, to find risk, or R, representing the 95% confidence level that cancer will result at the chemical uptake level modeled (EPA, 2017, 1999).

$$R = CDI*SL \quad (2)$$

Here, risk is set equal to CDI times the slope factor, SL. The slope factor is derived from laboratory experiments on chemical interactions and integrates relatively high doses of a chemical, compared to expected low doses in the environment (Subramaniam et al., 2006). For non-cancer impacts, to determine the potential hazard a comparison is made between the non-cancer reference dose (RfD), and the CDI. If the CDI is less than the RfD, the potential for hazard impact (or hazard quotient, HQ) is small.

$$HQ = CDI/RfD \quad (3)$$

Here CDI is the actual exposure level developed in the model (mg/kg/day) and RfD is the reference dose from peer reviewed studies. If, based on the screening HQ value, HQ < 1 then the exposure to the chemical poses no significant potential for adverse health effects, and further investigation may not be warranted. If the initial HQ estimate exceeds 1, then further investigation may be needed into the potential for adverse effects due to pollutant intake over time (EPA, 1999).

3. Results

3.1. Drinking water

In all iterations of the model, the risk from oral uptake of glacial water, even with lifetime consumption, is low (Table 1). Risk from consumption of meltwater is up to 4 orders of magnitude below screening level, suggesting insignificant risk within any timeframe. While the risk to children, or lifetime consumers, is marginally higher than the risk to adults, it is still below the screening level. These results indicate that we cannot expect to see cancer or non-cancer impacts to consumers of glacial water. This suggests that risk from ingestion of PCBs in glacial meltwater from Silvretta Glacier is negligible across uptake periods and body sizes.

3.2. Fish consumption

Fish consumption is the main driver of risk for both children and adults. This exposure pathway contributes the greatest risk to long term cancer and non-cancer hazard, assuming continued consumption over time. For children eating domestic fish as 6% of their yearly fish consumption, or 0.0013 kg/day (Statistical Office, 2017), out of water with a mean adjusted PCB concentration of 286 pg/L, cancer risks are above acceptable cancer screening level, indicating potential risk and the need for further assessment at an individual level. Increasing the PCB concentration to the upper limit concentration of 500 pg/L, and holding fish consumption steady, cancer risk is also above the screening level, with non-cancer hazards jointly indicated (Table 1). This would continue to be true for upper-limit concentration values of 500 pg/L, indicating a proportional increase of risk.

For adults with the mean fish consumption rate of 0.0013 kg/day and mean adjusted PCB concentration in meltwater of 286 pg/L, cancer risk is just above acceptable screening level for cancer, and below the non-cancer screening level, indicating minimal risk and that further study may not be necessary. Increasing PCB concentration values to 500 pg/L, adults remain slightly above the cancer screening level and below non-cancer values (Table 1).

The model indicates that even within a population-level screening analysis of glacial meltwater risk incorporating non-glacial sources, secondary emission of PCBs in glacial meltwater has the potential to lead to previously unexplored risks to human health. While the risk assessment thresholds used by the EPA screening-level model are not indicative of direct cancer rates in individuals, they indicate the need to enact the next step of the risk assessment framework, monitoring and mitigation at the local level.

4. Discussion

4.1. Drinking water

Due to its proximity to pollution sources and relatively high concentration of PCBs (Glüge et al., 2017; Miner et al., 2017; Pavlova et al., 2016), Silvretta Glacier represents the highest potential for risk to humans from POP reemission in glacial meltwater. This may suggest that interaction with glacial meltwater, topically or through drinking and inhalation, may have an adverse impact. However, we show that the human health risk from consumption, or physical interaction, with Silvretta meltwater is likely negligible in both adults and children, even with long-term interaction. Geographically isolated locations with very low glacial POP concentrations such as the Arctic could be expected to have even less risk from drinking glacial meltwater, in concordance with the latitudinal gradient from Europe to the Arctic (Garmash et al., 2013; Grannas et al., 2013). This kind of variable-constraining result is the overall goal of the screening-level tool, which was developed to identify and prioritize the regions or model variables critical for future study. Our risk assessment has accomplished this goal, iterating that the risk from direct uptake of POPs in glacial meltwater is negligible, and therefore should not be considered a priority for future studies on human health risk. Accordingly, both our exposure and screening level model suggest that in the northern hemisphere the impact of human consumption of glacial meltwater containing the level of PCBs released within the past 10 years is insignificant in any timeframe.

4.2. Fish consumption

While fish consumption rates are relatively low in Switzerland (Statistical Office, 2017), our model suggests caution must be exercised by populations that frequently consume fish from glacial watersheds, due to the high potential for PCB bioaccumulation. The toxicity and bioaccumulation potential of PCBs make the chemical class a specific concern for biomagnification in both large and long lived species and may necessitate further fish population monitoring (Czub and McLachlan, 2004a, 2004b; Office of Environmental Health Hazard Assessment, 2012). While the Silvretta watershed PCB concentrations are significantly lower than those seen in industrial watersheds, the relatively recent addition of glacially derived chemicals may increase the risk of cancer and non-cancer impacts in resident populations (EPA, 2001b; Birnbaum, 2013). This emergent risk must be incorporated into any population risk profiles, additionally necessitating further research into the toxicology of glacial meltwater. Populations that rely on fish consumption from glaciated watersheds to meet their energy needs, including indigenous populations in the Arctic, may have proportionally higher risk and should be assessed individually. As glacier melt within the Swiss Alps increases under climate warming, the release of POPs in glacial meltwater may fluctuate and should be monitored. Risk can be minimized on a local level in some situations by monitoring fish bioaccumulation in watersheds of concern, in combination with monitoring chemical release from glaciers long-term.

While this risk assessment incorporates PCB uptake, additional chemicals that were not investigated here, but are resident in glacial meltwater from similar watersheds, may increase risk. For example, samples taken from glaciers in the Italian Alps have identified the presence of hexachlorobenzene (HCB), hexachlorahexene (HCH), dichlorodiphenyltrichloroethane (DDT) and current use pesticides (Ferrario et al., 2017; Villa et al., 2006, 2003; Wu et al., 2017). Including multiple chemicals in a sampling protocol may further increase the risk to above screening level, when considered as mixtures. The combination of changing glacial flow rates and unprecedented warming will also need to be monitored. Further monitoring of new glacial meltwater inputs and residual concentrations will ensure the health of residents and visitors to these systems.

4.3. Natural variability

Uncertainty inherent to the natural ecosystem creates difficulties in direct modeling of chemical fluctuations within glacial melt and hydrology. Application of the risk assessment methodology to a glacial system requires an understanding of glacial morphology, watershed structure, and human uptake rates and routes (Bogdal et al., 2010; Grannas et al., 2013). Glacial ice, firn and snow all have the ability to act as a POP reservoir, but with respectively decreasing residence times and melt timeframes (Cuffey and Patterson, 2010; Davidson et al., 2003; Jansson et al., 2003). Warming temperatures and changes in precipitation type can increase glacial melt chemical flux and opportunities for downstream POP distribution with the length, thickness and response rate of a glacier directly impacting when pollutant release will occur (Cuffey and Patterson, 2010; Hock, 2003; Hooke, 2005; Jansson et al., 2003). Complications in modeling future release of pollutants to the granularity needed for risk assessment emerge from unknown rates of atmospheric warming and melt patterns combined with unknown chemical positionality within the glacier and incorporation into the watershed (Farinotti et al., 2009; McNeill et al., 2012; Steinlin et al., 2016). Therefore, variability in glacial melt over time limits the ability to predict future POP concentrations accurately under a warming climate.

We attempted to constrain this variability by utilizing both the maximum and the average PCB concentrations from 2 different sampling techniques over two years. All of the concentration data are from the mixed-stream source that most closely represent local lakes and streams in composition (P7-Fig. 1) (Finkel, 2014; Quadroni and Bettinetti, 2017; Schmid et al., 2012). These values are within the same magnitude of POP concentration data taken across the Alps range, integrating numerous sampling months over multiple years (Bogdal et al., 2009; Ferrario et al., 2017; Schmid et al., 2011; Villa et al., 2006). This allowed us to identify the median and end member pollution scenarios that may be developing within glaciated watersheds. While glacial melt may not occur year-round in some ecosystems, bioaccumulation in fish will reflect long term concentrations, which are growing steadily in some glacial watersheds (Miner et al., 2018).

The inherent variability of the linked glacial-atmospheric system leads to the possibility of non-linear melt over an abbreviated period which could subsequently increase variability of chemical concentration within the watershed. This potential scenario has not been incorporated in our work, as glacial flow models of Silvretta Glacier do not currently forecast non-linear melt (Steinlin et al., 2015).

In addition to the complications of modeling glacial POP distribution, the application of a screening-level model that does not utilize individual consumption, body weight or lifetime data, but instead uses the population mean, requires certain assumptions. For this linear model we utilize consumption, body weight and exposure duration data that take the population mean, in line with the established EPA methodology (EPA, 2001a, 2001d). Though not specific to individuals, standardized consumption rates for fish and water are considered typical for screening level risk assessments, and are put forward by the EPA and further refined by country statistics (EPA, 2001a, 1999). These values may not adequately represent the natural biological variation in individuals, or represent vulnerable populations with greater pollutant susceptibility (Frey, 1992; Gallagher et al., 2015). To this end, we conducted a sensitivity analysis that indicated that a departure of ± 20% from suggested values would not move risk to the other side of the risk threshold, for either positive or negative risk. However, it is appropriate that the results of this screening model be viewed as an indication of the need for further study since standard thresholds have been breached. Recent studies of PCBs in glacially derived water sources from the Italian Alps have similarly identified potential human health risks from bioaccumulation of PCBs in fish (Bettinetti et al., 2016). Our study corroborates these findings while integrating additional human consumption dynamics on a population scale.

5. Conclusion

In selected glacial watersheds throughout the world, the release of POPs from melting glaciers may be counteracting the trend towards POP reduction in the global environment. We show that the risk from glacial meltwater release of pollutants is predominantly through bioaccumulation, and that the implications of POP movement through the food chain and into the human system must be investigated further. These further analyses should incorporate site-specific details and vulnerabilities, prioritizing regions where glacial meltwater is used or where glacial watersheds provide food sources directly to the population, especially in the southern hemisphere. While the risk associated with consumption of glacial meltwater is small and may continue to decrease, the impact of legacy pollutants in glacial systems is ongoing and must be prioritized in future research.

Acknowledgements

We thank Glenn Rice from the US EPA, without whom this research would not have been possible. We also researchers from the Climate Change Institute for their ongoing support. KR Miner was supported by the Robert and Patrica Switzer Foundation, ASEE SMART Scholarshipand National Science Foundation grant DGE- 1144423.

Supporting information

Further details on model parameters, values and model layout available.

The authors have no competing interests to declare.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2018.09.066.

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